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Analysis of magnetic nanoparticles using second harmonic responses

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ABSTRACT

Magnetic particle imaging (MPI) is a new medical imaging technology with a number of potential applications. It is based on utilizing the non-linear magnetization response for magnetic nanoparticles (MNPs). We have proposed a method to improve the detection sensitivity for the magnetization of MNPs, and their imaging technique, based on the detection of a second harmonic of the response using a high Tc SQUID. The advantage of using the second harmonic is that the response can be measured even with small amplitudes. In this paper, superparamagnetic MNPs with different diameters and different compositions of materials were analyzed by this method. In the MNPs, Resovist was found to be almost one order of magnitude larger than that of the other MNPs tested. The magnetic moments of the MNPs were estimated by fitting with the Langevin function to obtain the magnetic moments m of Resovist of 3.62×10^{-18} [J/T]. The magnetic moment m of Resovist was 7.4 times larger than the other MNPs.

1. Introduction

Magnetic particle imaging (MPI) technic has been introduced by Gleich and Weizenecker, which is based on utilizing the non-linear magnetization response M for the detection of magnetic nanoparticles (MNPs) [1]. The magnetization response contains not only the fundamental excitation frequency, but also its harmonics, when an ac excitation magnetic field H_{ac} is applied. A number of magnetic detection methods have been developed to determine the mass of MNP for different applications such as immunoassays [2,3]. In MNP detection and the MPI technique, the most commonly employed method is measurement of the odd harmonics of the M response. We have proposed a method to improve the detection sensitivity of the M of MNPs, and thus MNP imaging techniques based on the detection of the second harmonic of the response [4]. Upon the application of H_{ac} along with an additional dc bias field, the second harmonic of M reaches a maximum due to nonlinearity in the M–H characteristics. If the amplitude of H_{ac} is relatively lower, the second harmonic will be the strongest [5,6]. In this paper, several superparamagnetic MNP samples of different particle diameters and different material compositions were evaluated utilizing the second harmonic response method.

2. Evaluation of magnetic nanoparticles

2.1. Experimental setup

Fig. 1 illustrates the experimental setup for the evaluation of the MNPs [7]. The setup consists, in essence, of a dc field coil, an ac modulation coil, a differential detection coil, an input coil, and an RF high-T_c superconducting quantum interference device (SQUID). The SQUID was used as an ultra-low noise amplifier. All the coils were arranged in a collinear configuration, except for the input coil. The differential detection coils each had 240 turns. The input coil had 1000 turns, with an internal diameter of 30 mm and a length of 21 mm. The coil parameters, including the measured resistances and impedances, are listed in Table 1. A thin-film $Y_1B_2C_3O_{7-y}$ SQUID with a step-edge junction was placed at the center of the input coil. The SQUID and input coil were cooled by liquid nitrogen in a 30 L aluminum Dewar, which was surrounded by a three-layer, 2-mm-thick mu-metal cylinder. The mutual inductance measured with the SQUID was 16 nH. The signal to noise ratio (SNR) of the SQUID was previously measured to be 3.6 times higher than that of a semiconductor amplifier [7].

2.2. Evaluation of MNPs

Two types of superparamagnetic MNP samples were prepared, using MF and Resovist, based on Fe_3O_4, and Fe_2O_3, respectively. These

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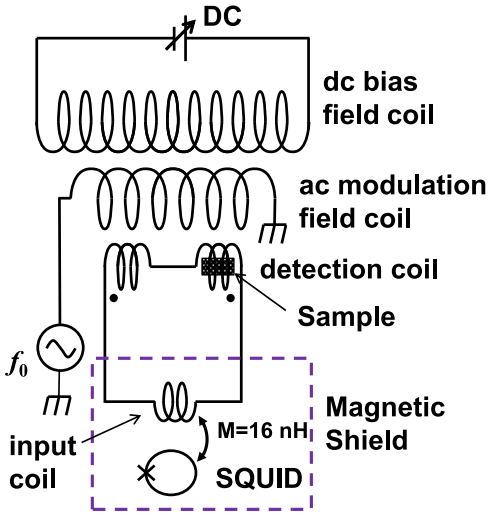

Fig. 1. Experimental setup for MNPs evaluation.

Table 1
Coil parameters.

	No. of Turns	Length [mm]	Wire Dia. [mm]	Induct. [mH]	DC Res. [Ω]	H/I [(mT/μ_0)/A]
dc bias field coil	1040	100	0.9	34.8	6.5	10.6
ac mod. field coil	541	120	0.4	0.7	4.2	6.8
detection coil	240 +240	12+12	0.2	0.8	4.8 +4.8	–
input coil	1000	21	0.4	26.4	2.3 @ 77 K	–

Table 2
Details of prepared MNP samples.

Sample name	Core	Fe [mg/ml]	Dia. [nm]
MF-30	Fe_3O_4	8	36.3
MF-60		8	52.1
MF-90		8	96.7
Resovist (original conc.)	Fe_2O_3	27.8	60

are hydrophilic colloidal solutions of iron oxide coated with carboxydextran, with particle sizes ranging from 30 nm to 100 nm. Details of the prepared MNP samples are listed in Table 2. The concentration of the original Resovist solution was adjusted to 7.94 mg/ml with deionized water to obtain a similar concentration to that of the MF solutions.

The static field generated by the dc bias field coils reached amplitude values of up to ± 21.2 mT/μ_0, with a 5 kHz ac modulation field of up to 2.43 $mT_{p\text{-}p}/\mu_0$ applied to the sample. The sample rod containing a small vessel with inner dimensions of ϕ5×6 mm was placed in the bore of the detection coil. The vessel was filled with 70 μl of an MNP sample, and placed so that the sample was in the middle of one side of the differential detection coil. The output signals from the SQUID electronics were measured by a dynamic signal analyzer (35670A, Agilent Technologies).

The dependence of the second harmonic peaks of each MF (Fe_3O_4) sample on the dc bias field H_{dc} is shown in Fig. 2(a). Two peaks are observed in the region of H_{dc}= ± 5 to 10 mT/μ_0. The MF-90 (ϕ96.7 nm) sample displays the largest peak value, with peak values of the other samples (ϕ52.1 nm, ϕ36.3 nm) decreasing with their particle size. Conversely, the third harmonic response shown in Fig. 2(b) shows a peak at H_{dc}=0, which corresponds to the FFP on

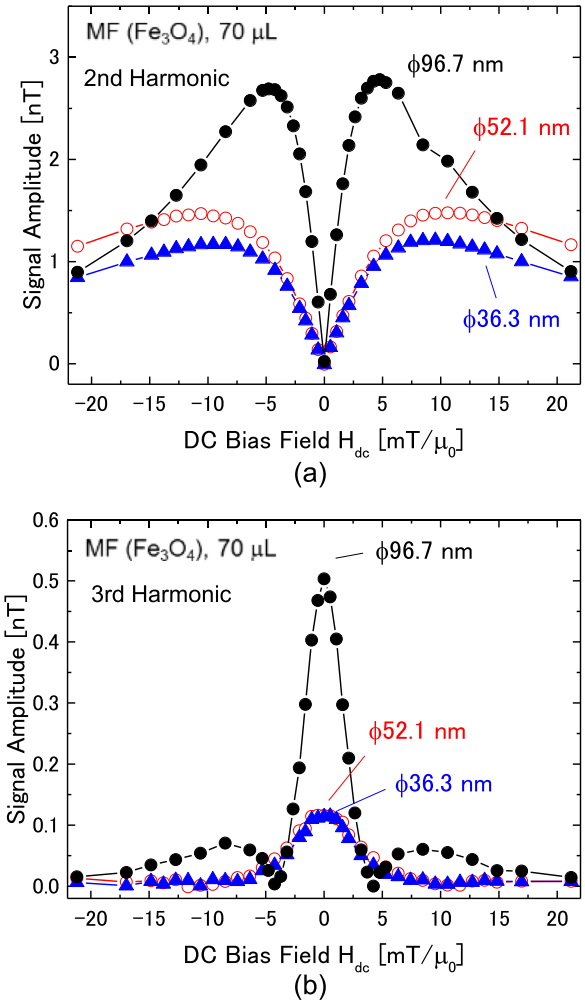

Fig. 2. Dependence of the harmonic response of each MF sample on the dc bias field H_{dc}: (a) The second harmonic peaks; (b) The third harmonic peaks.

the *M-H* curve. The peak value of MF-90 is again the largest; however, it is one order of magnitude smaller than that of the second harmonic.

The diluted Resovist sample (Fe_2O_3, 7.94 mg/ml) was evaluated, and the dependence of the second harmonic peaks on the dc bias field H_{dc} is shown in Fig. 3. Two peaks are observed in a similar manner to the MF samples. However, the peak value is 24 times larger than that of the MF-60 sample with a similar particle diameter to Resovist, which is

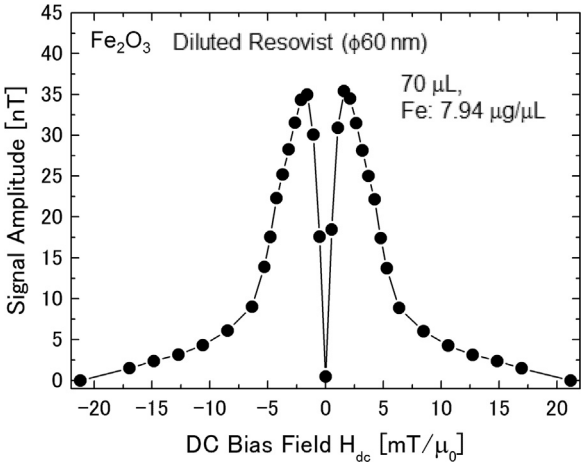

Fig. 3. Dependence of the second harmonic response of Resovist (ϕ60 nm) on the dc bias field H_{dc}.T.